Senior Thesis

Distribution of Na and Ca in brines of the Clinton sandstone of Ohio

by
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ABSTRACT

The Na and Ca concentrations of 45 brine samples from the Clinton sandstones in eastern Ohio are the result of mixing of two brine components in the subsurface. These concentrations were subsequently altered by dilution with groundwater or meteoric water. The two brine types are: a Na-rich brine and a Ca-rich brine. The abundance of the Na-rich brine slightly decreases with depth in the Clinton, while the abundance of the Ca-rich brine slightly increases with depth, but both are virtually constant.

Four Na-rich, low-Ca anomalies exist in eastern Ohio. Three of the four anomalies correlate with known basement faults in eastern Ohio. Therefore, this study supports the conclusion that mixing and dilution account for the observed chemical compositions of brines in the Clinton sandstones of eastern Ohio.

INTRODUCTION

Chemical compositions of subsurface brines may vary due to mixing of two or more different brine types and by dilution of the resulting mixture with meteoric water. Variations in chemical composition of oilfield brines may also be caused by contamination with fresh water used in hydrofracturing of hydrocarbon-bearing strata or by faulty well casings and seals that permit groundwater to leak into producing wells. Additionally, brines may be contaminated with meteoric water in storage tanks or as a result of inappropriate sampling techniques (FAURE *et al.*, 1988).

The purpose of this study is to test the hypothesis that the variations in the Na and Ca concentrations of oilfield brines of the Clinton sandstones in eastern Ohio are the result of mixing of two distinctly different brine components followed by dilution of the resulting mixture with varying amounts of meteoric water.
In addition, this study explores the relationship between anomalies of Na and Ca concentrations and faults in the basement rocks of eastern Ohio. The data used in this study come from 45 wells whose geographic locations are indicated in Figure 1.

A positive correlation between elemental anomalies in oilfield brines and basement faults could lead to the discovery of new faults. This information is useful to the petroleum industry, because the identification of zones of basement faulting during basin filling provides a framework for understanding sediment distribution patterns and lithologic variations in oil and gas reservoirs.

Previous brine studies in Ohio that have been based on chemical compositions are done by STOUT *et al.* (1932), LAMBORN (1952), THOMPSON (1973) STITH (1979), and PARSONS (1982). The most recent study is by LOWRY (1986) also on brines of the Clinton sandstone.

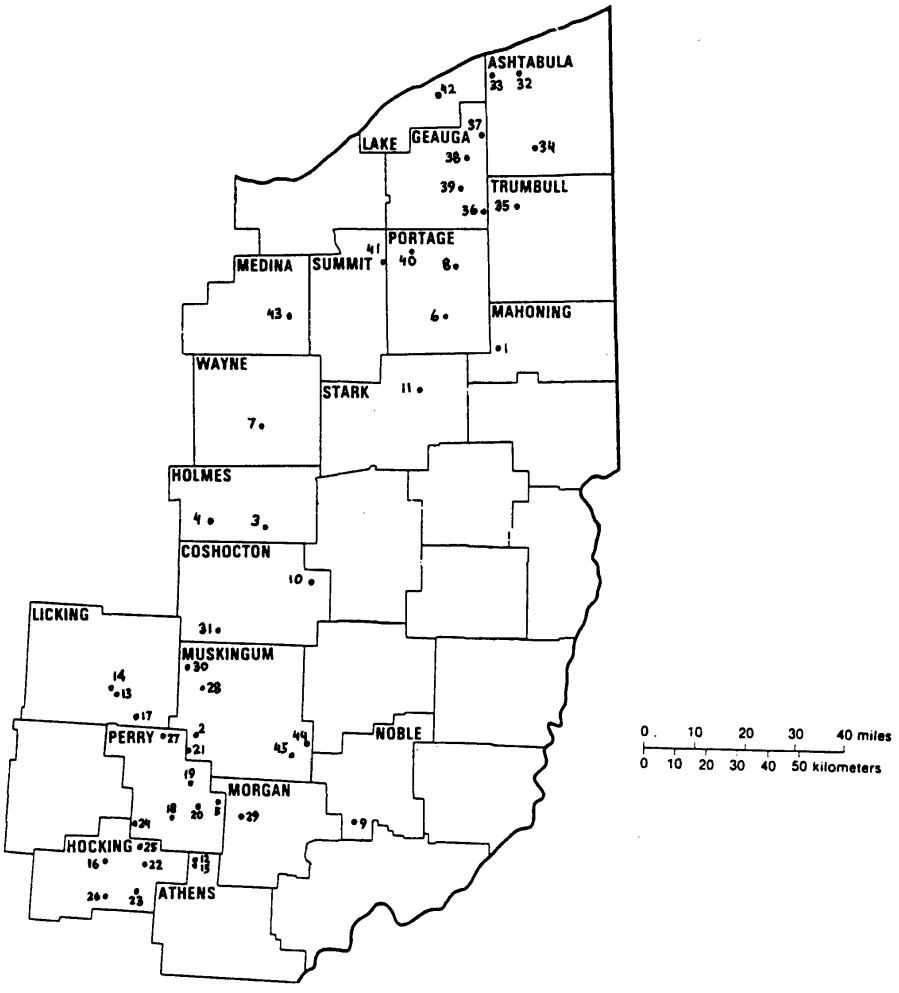

Figure 1. Map of eastern Ohio showing the locations of the 45 gas and oil wells from which brine samples were analyzed.

THE CLINTON SANDSTONE

The Clinton sandstone is Early Silurian in age and is a major gas and oil producer in Ohio. The Clinton sandstone was named by drillers in 1887, but the United States Geologic Survey recognizes these beds as the Albion Formation (LOWRY, 1986). Despite this fact, the Ohio Geologic Survey accepts the term Clinton. Information about the Clinton sandstone is obtained from studying core samples and geophysical logs, because the Clinton does not outcrop anywhere in Ohio.

The Clinton sandstone is further subdivided in an informal basis by using the drillers' names. The upper tongue of the sandstone is the drillers' Clinton. The part intertonguing with the lower tongue of the Cabot Head Shale is referred to as the White Clinton, because it is generally white. The part intertonguing with the upper tongue of the Cabot Head Shale is called the Stray Clinton, due to its erratic distribution. The central sandstone between the White and Stray Clinton is the Red Clinton, as it is commonly red and is so identified by drillers (KNIGHT, 1969).

Petrology

The lowermost White Clinton is a gray, very fine-grained grained quartz sandstone and a coarse- to medium-grained quartz siltstone. Silica is the cementing material, although some calcite and clay minerals are present. The grains are generally subrounded. The upper White Clinton consists of a white to very light-gray quartz sandstone which is medium- to fine-grained. The grains are angular to subrounded, and silica is the cementing material.

The Red Clinton is a mottled red and gray color. It is very fine- to fine-grained sandstone, and a coarse grained quartz siltstone. The cementing materials are silica and hematite, which gives the sandstone its red color. Sand and silt grains are

subangular to subrounded, becoming generally smaller and less angular upward.

The Stray Clinton consists of a white to light-gray quartz siltstone, which is coarse- to fine-grained, and a very fine-grained quartz sandstone. The siltstone is predominant. The cementing material is silica; some calcite and clay minerals are also present. The grains are subrounded to subangular, with angularity and grain size decreasing upward (KNIGHT, 1969).

Stratigraphic Position

Figure 2 is a partial stratigraphic section of the study area. The Clinton sandstone lies directly above the Ordovician Queenston Shale, called Medina by drillers. Above the Clinton sandstone is the Packer Shell, a drillers' name for a coarsely crystalline limestone, which is equivalent to the Brassfield limestone of the Ohio Geologic Survey. The Paleozoic rocks of eastern Ohio form a homocline that dips in a southeast direction.

	Group	Formation or principal lithology of Ohio Geologic Survey	Drillers' terms
S	Bass Islands	Dolomite	
I	Salina	Dolomite	Big Lime
L	Lockport	Dolomite	
U		Shale, limestone, dolomite	Packer Shell
R		Thorold Sandstone	Stray "Clinton"
I	Albion	Grimsby Sandstone	Red or 1st "Clinton"
A		Cabot Haed Shale	White or 2nd "Clinton"
N		Whirlpool Sandstone	
ORDOVICIAN		Queenston Shale, Reedsville Shale	Red Medina

Figure 2. Generalized stratigraphic section for eastern Ohio (Modified after BREEN *et al.*, 1985).

PRINCIPLES OF MIXING

Mixing Triangles

When a brine of constant composition is progressively diluted with groundwater or meteoric surface water, the concentrations of any two conservative elements from the brine samples form a straight line that is directed toward the origin. When two brines of differing composition mix in the subsurface, and are diluted by groundwater or meteoric surface water, the resulting points are scattered within a triangle of mixing (FAURE *et al.*, 1988). Two of the three sides of the triangle are formed by extending a line from the origin, O, to each of the different brine types, A and B for example. The third line is the "mixing line", which connects A and B as shown in Figure 1. The mixing triangle can be used to determine: 1) the concentration of component A or B in an individual sample; 2) the degree of dilution of that sample; and 3) the concentrations of the elements under study in the undiluted brine.

Concentrations of Brine Components

To determine the extent of mixing of an individual sample, a line is extrapolated from the origin that passes through the sample point to the mixing line. For example, in Figure 3 a line has been drawn from the origin, through sample point S and onto the mixing line, terminating at point C. To determine the concentration of brine A in this sample, the distance along the mixing line from C to B is divided by the total length of the mixing line, from A to B. The concentration of brine A in a given sample is denoted f_A. The concentration of brine B, denoted f_B, is calculated in the same fashion, by dividing the length from A to C by the total length of the mixing line. If f_A is known, f_B can be calculated by the

simple equation:

$$f_B = 1 - f_A$$

Because of this relationship, f_B was not calculated to avoid redundancy.

Dilution

The dilution of a given sample, denoted f_D, can be determined by dividing the measured distance of the portion of the extrapolated line between the sample point and the mixing line by the total length of the extrapolated line. For example, in Figure 3, the distance from S to C is divided by the distance from O to C to obtain f_D.

Elemental Concentrations

The removal of the effects of dilution from a given sample produces a point that lies at the intersection of the mixing line and the line that passes through the sample point. In Figure 3, this location is at point C for sample S. The undiluted elemental concentrations under consideration can be read from the coordinates of point C.

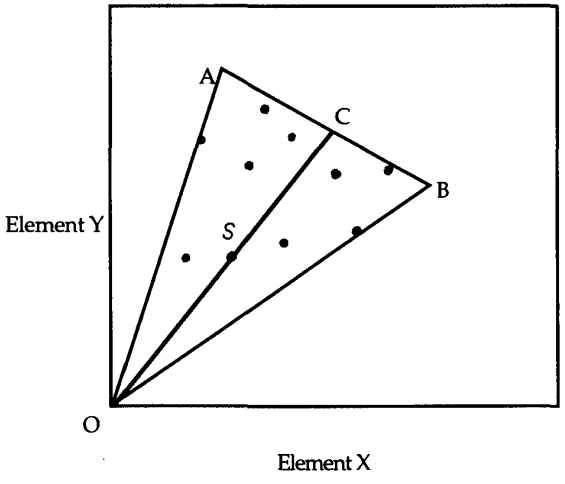

Figure 3. Sample plot of the concentrations of Na and Ca from brine samples enclosed within a mixing triangle. The diagram shows mixtures of two brine types A and B which have been diluted with meteoric water.

METHODS

The Na and Ca concentrations for 45 brine samples obtained from the Clinton sandstone are shown in Table 1 of the Appendix. The concentration of Na was plotted versus the concentration of Ca for these samples. The resulting array of points was enclosed within a mixing triangle. A preliminary evaluation indicated that samples 1 and 11 were anomalous, suggesting that they may have been affected by a third brine. Therefore, a new mixing triangle was drawn that excluded samples 1 and 11.

After the mixing triangle was drawn, f_A, f_D, and undiluted concentrations of Na and Ca were determined for each sample (excluding 1 and 11). Contour maps were drawn for the raw data for Na and Ca concentrations. Next, contour maps were created for f_A and f_D and for the concentrations of Na and Ca with the effects of dilution removed.

Finally, in a test for a relationship between brine composition and depth, well depth was plotted against f_A. Superimposed over this plot are increments of one standard deviation (67% probability). Wells in Muskingum, Noble, Perry, Hocking, and Athens counties were excluded from this statistical manipulation due to the anomalies which exist in these counties, which may tend to skew the true relationship between brine composition and depth.

RESULTS

Raw Data

The raw Na and Ca concentrations of the brine samples from eastern Ohio are listed in Table 1 of the Appendix. The concentrations of both elements increase eastward as shown in Figures 4 and 5, respectively. This is expected when one

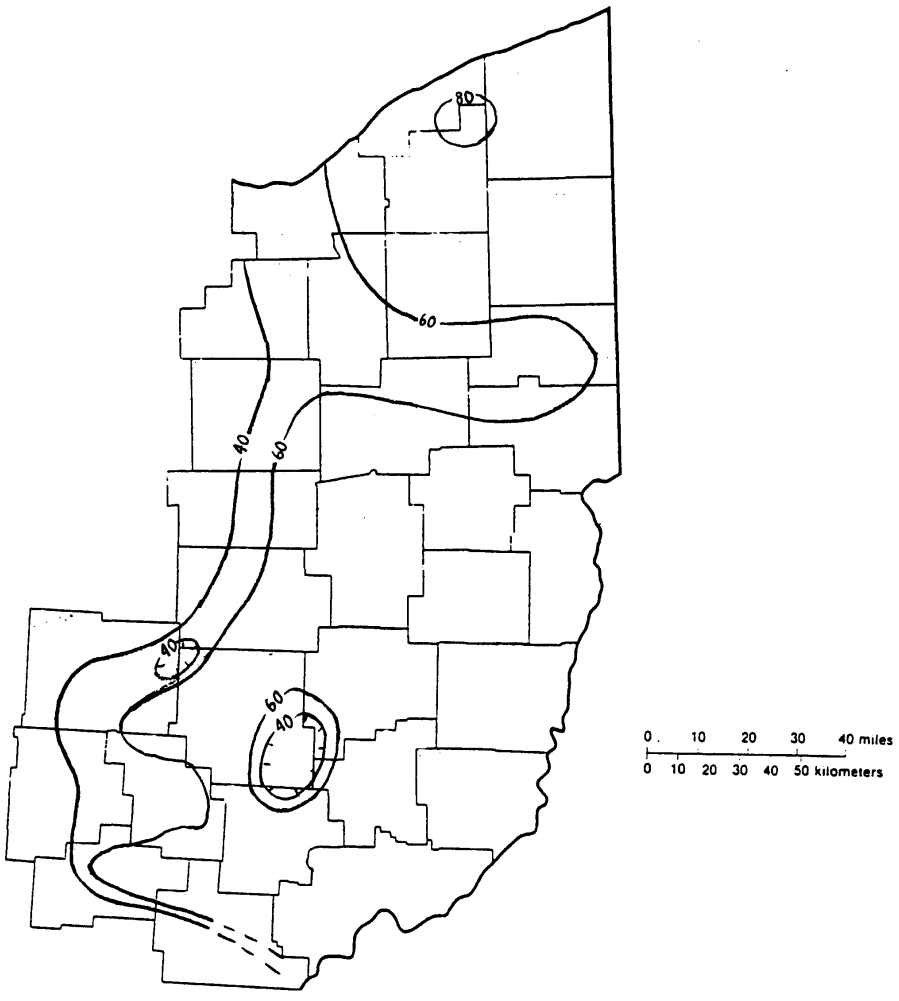

Figure 4. Contour map showing the westerly decrease in Na concentrations in the Clinton sandstones caused by dilution of the brines by recharge of meteroic water from the surface. Contour interval is 20,000 mg/L.

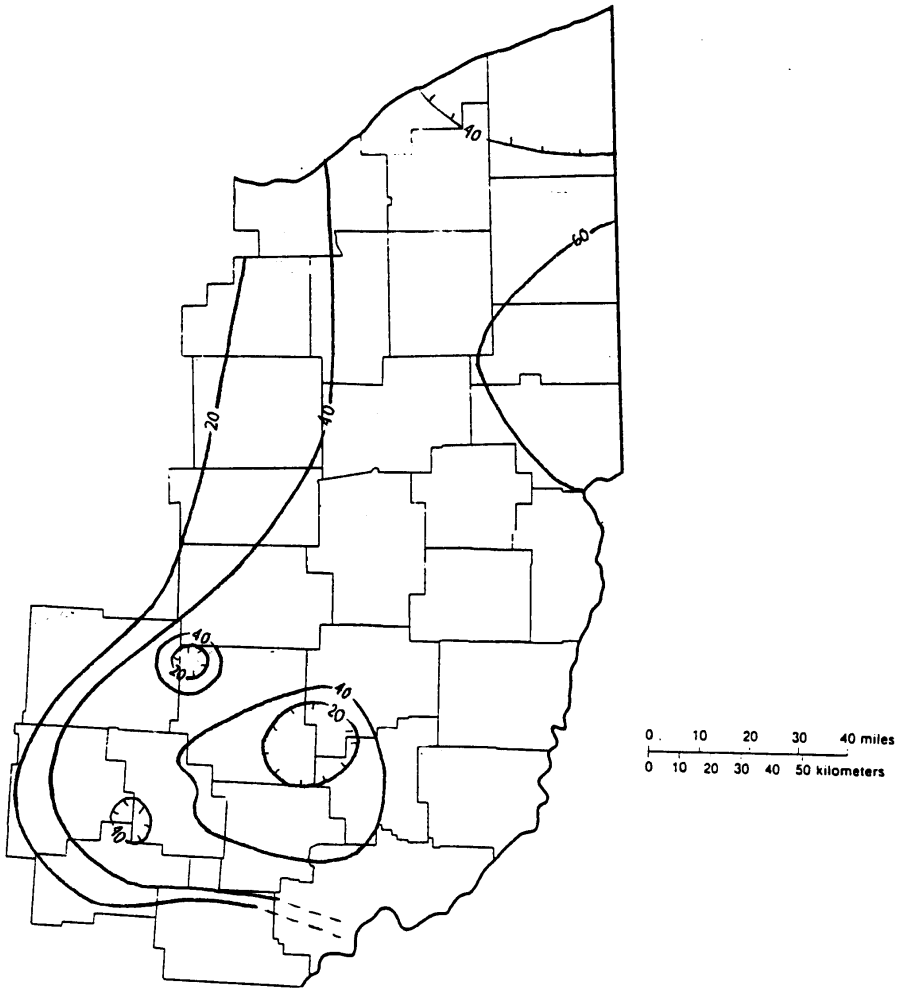

Figure 5. Contour map showing the westerly decrease in Ca concentrations in the Clinton sandstones caused by dilution of the brines by recharge of meteroic water from the surface. Contour interval is 20,000 mg/L.

considers the geology of eastern Ohio. The Clinton sandstone dips to the east and southeast, and is at its shallowest depth where the Na and Ca concentrations are lowest. Therefore, it is expected that brines in the western part of this study are most diluted by meteoric water. Because of this expectation, the amount of dilution was determined for each sample.

Dilution

Amounts of dilution were determined for each sample, and are listed in Table 2 of the Appendix. As expected, dilution (f_D) was higher in the western part of the study area, as seen in Figure 6. Some high-dilution anomalies exist in Muskingum, Perry, and Hocking counties.

Undiluted brines

After the effects of dilution were removed from the brine samples the variation from the Na and Ca concentrations was reduced, which can be seen in Figures 7 and 8, respectively. Most Na concentrations are between 60,000 and 80,000 mg/L, and most Ca concentrations fall between 40,000 and 60,000 mg/L. These concentrations are given in Table 2 of the Appendix.

A number of anomalies exist with high-Na and low-Ca concentrations. These occur in northeast Ohio in Ashtabula, Geauga, and Lake counties, and in southeast Ohio in Noble, Muskingum, and Hocking Counties. Sample 30 in Muskingum County has the most extreme values, with a Na concentration of 89,000 mg/L and a Ca concentration of 14,000 mg/L.

Abundance of Brine Components

The mixing triangle identifies two hypothetical brine components labeled A and B. Brine A is Na-rich, having a Na concentration of 89,400 mg/L and a Ca

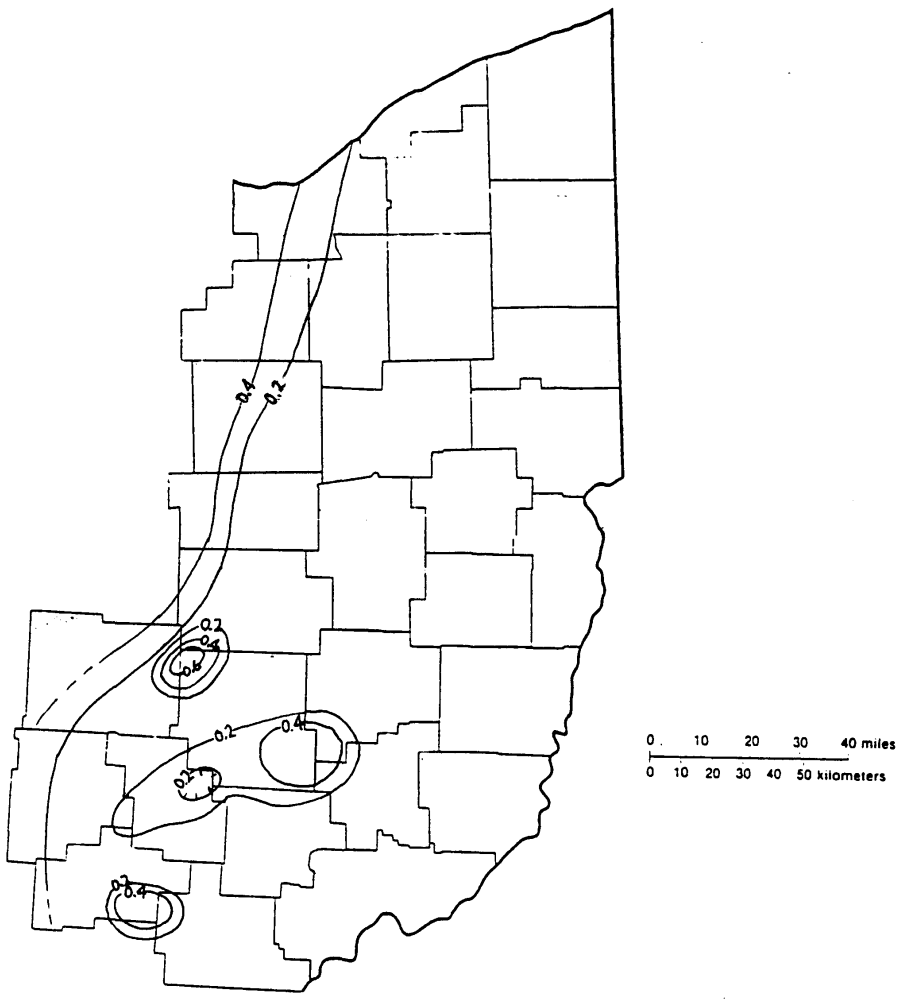

Figure 6. Contour map showing the westerly increase of the dilution (f_D) of brines in the Clinton sandstones. Significant anomalies exist in Muskingum, Perry, and Hocking counties. Contour interval is 0.2.

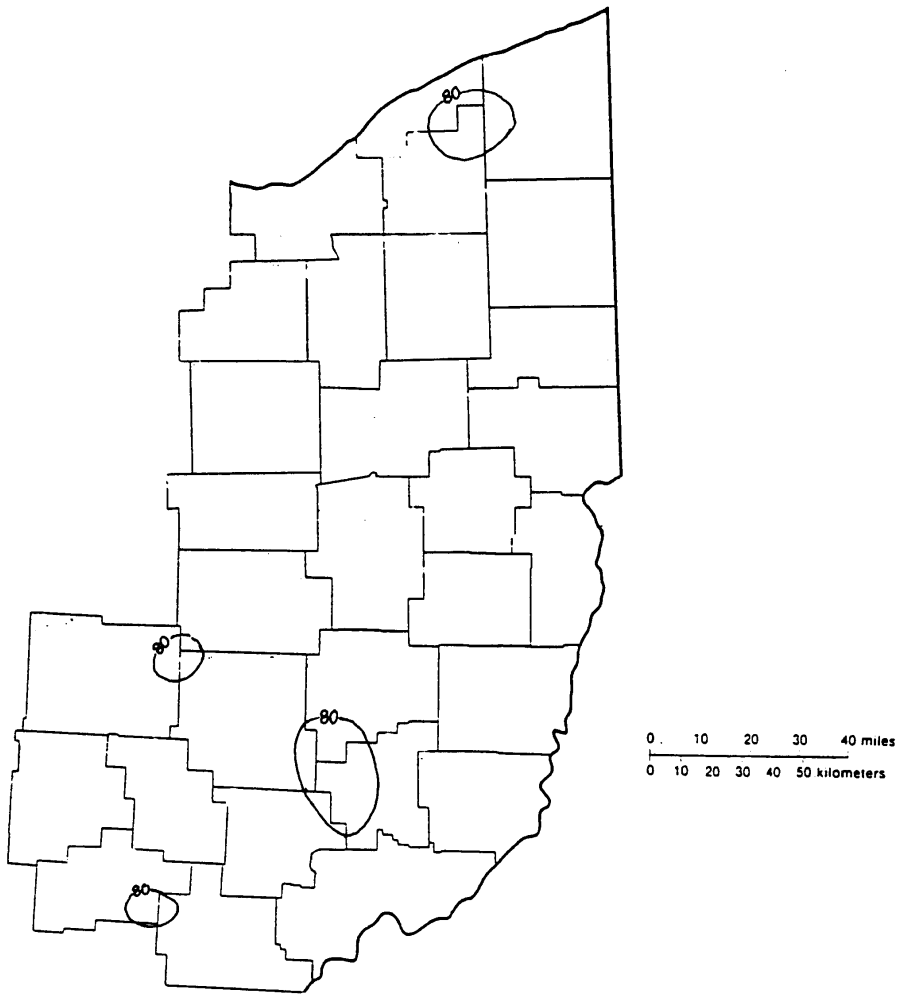

Figure 7. Contour map showing the Na concentrations (> 60 but < 80 mg/L) with the effects of dilution removed in the Clinton sandstones. Significant positive anomalies exist in Muskingum, Noble, and Hocking counties. Contour interval is 20,000 mg/L.

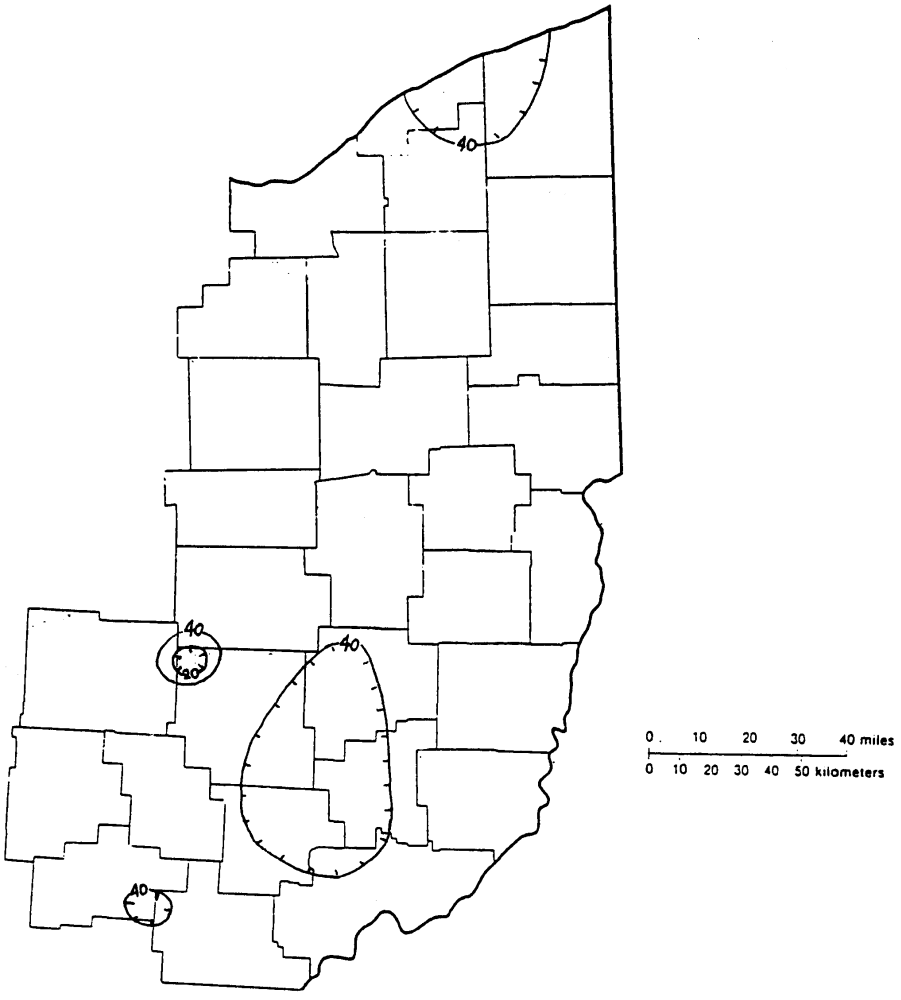

Figure 8. Contour map showing the Ca concentrations (> 40 but < 60 mg/L) with the effects of dilution removed in the Clinton sandstones. Significant anomalies existin Muskingum, Noble, Morgan, and Hocking counties. Contour interval is 20,000 mg/L.

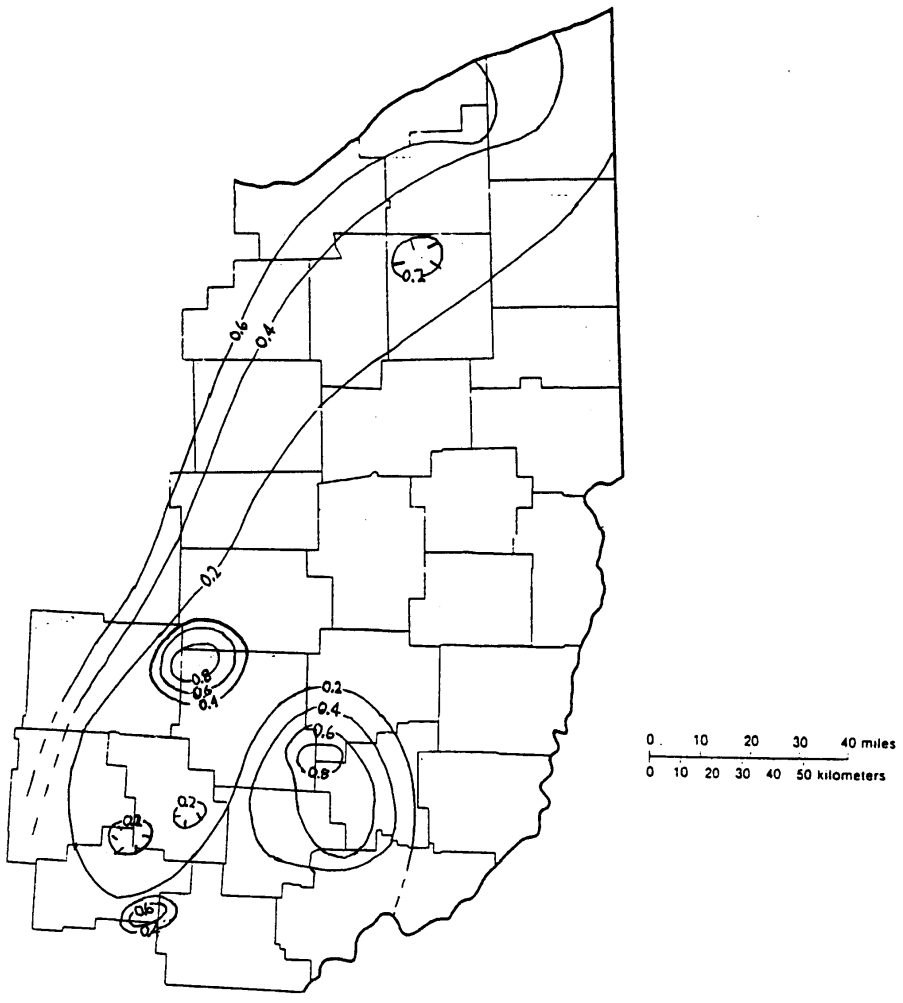

Figure 9. Contour map showing the westerly increase in the abundance of the Na-rich brine (f_A) in the Clinton sandstones. Significant anomalies exist in Muskingum, Noble, Morgan, and Hocking counties. Contour interval is 0.2.

concentration of 13,500 mg/L. Brine B is Ca-rich, having Na and Ca concentrations of 65,300 mg/L and 57,300 mg/L respectively.

In Figure 9 the concentrations of the Na-rich brine, denoted f_A, can be seen. In general, f_A decreases going eastward in the region. A number of anomalies do exist, however. High f_A values are found in Muskingum, Noble, and Hocking counties. The f values for all brine samples are displayed in Table 2 of the Appendix.

Relation between f_A and Depth

The depth of each well from which a brine sample was taken is listed in Table 2 of the Appendix. Depth was plotted versus f_A in Figure 8, producing a wide scattering of points. The data was condensed by dividing the total range of depth

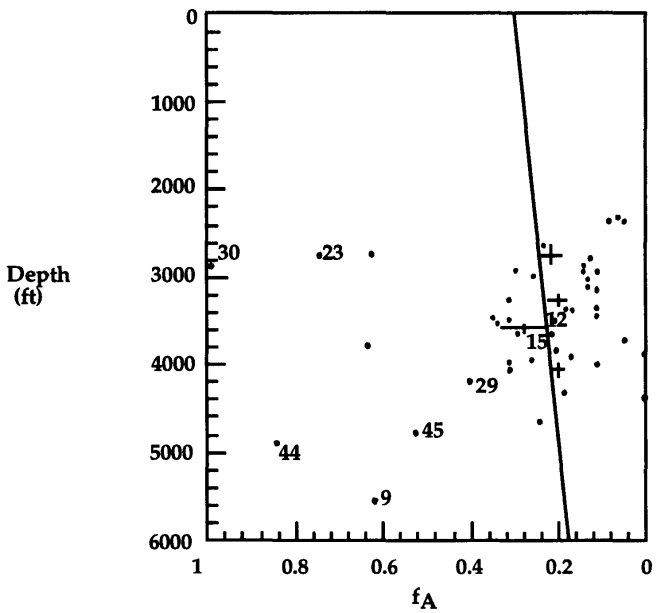

Figure 8. Relation between the abundance of the Na-rich component A and depth within the Clinton sandstones. Samples 9, 12, 15, 23, 29, 30, 44, 45 are anomalously enriched in Na, were not considered in the placement of the best-fit line.

into four intervals, and computing the average f_A and depth values for each interval, which produced four points. These points were enlarged by one standard deviation in each interval, thus indicating a confidence interval of 66%. The ranges of the confidence intervals, in addition to the mean and standard deviation of each depth interval are listed in Table 3 of the Appendix. The four points were superimposed over the scattering of points on Figure 8. A best-fit line drawn through these four points is nearly vertical, showing that f_A is virtually independent of depth.

DISCUSSION

Ca and Na Concentrations

Concentrations of undiluted Ca and Na in eastern Ohio fall within a narrow range, as mentioned before, with the exception of a few anomalies. These anomalies occur in approximately the same locations for both elements, which can be seen by comparing Figures 2 and 3. I think that it is likely that these anomalies are related to fault zones in the eastern half of the state.

There are two zones of major basement structures in eastern Ohio. The Burning Springs Anticline and Cambridge Arch (BS-C Fault Zone) define a narrow zone of north-northwest-trending Paleozoic faults, with a length of at least 160 miles (250 km), and possibly as much as 220 miles (350 km). In addition, the Highlandtown-Middleburg Fault Zone (H-M Fault Zone) extends 100 miles (160 km) northwest from Pennsylvania to Cleveland (ROOT and MARTIN, 1995). Figure 9 shows the locations of these major basement structures with superimposed contours of undiluted high-Na and low-Ca concentrations.

An examination of this figure shows a striking correlation between brine

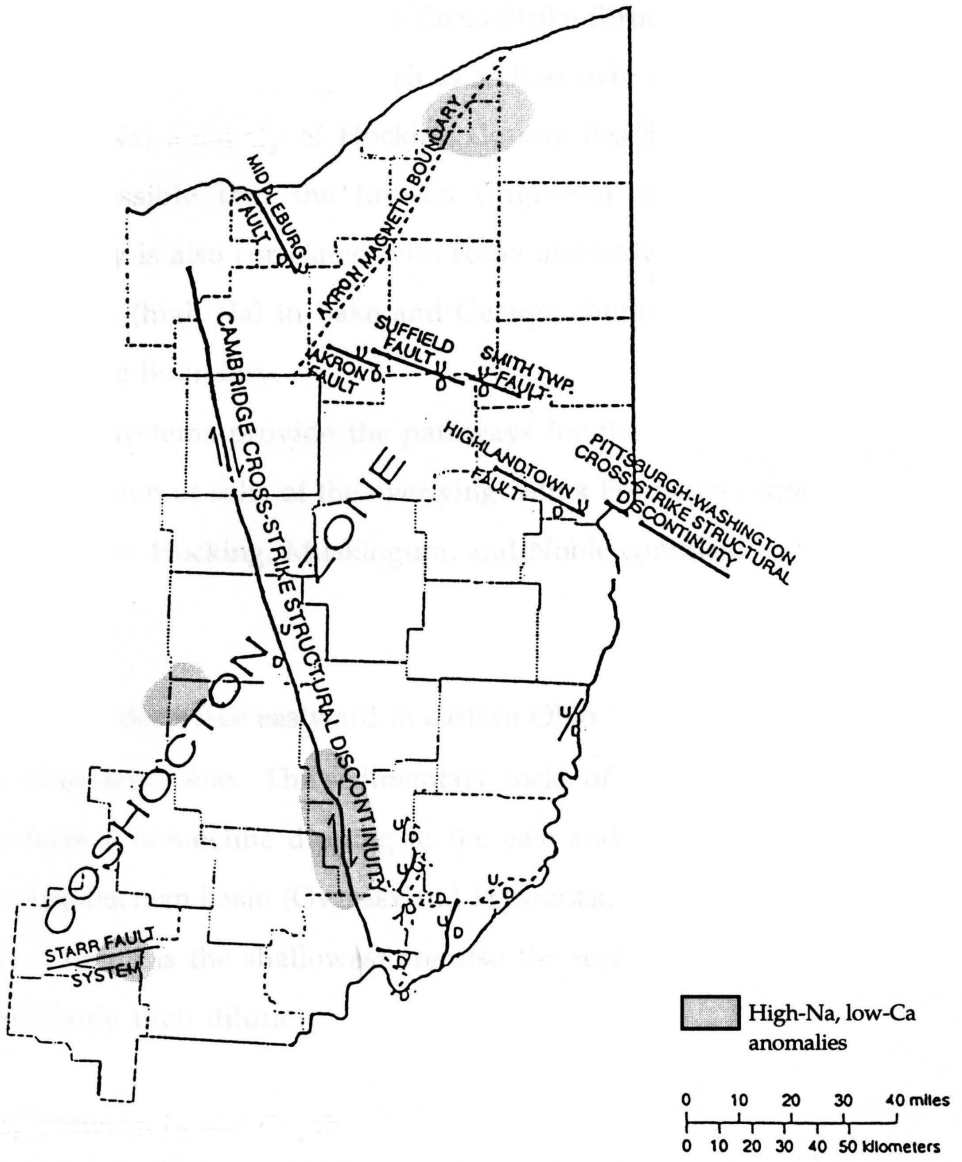

Figure 9. Map showing the relation of major basement structures in eastern Ohio to high-Na, low-Ca anomalies in brines of the Clinton sandstone. Modified from Root and Martin (1995).

anomalies and faults. The low-Ca (high-Na) anomaly of Morgan and Noble counties lies directly on the Cambridge Cross-Strike Structural Discontinuity, more commonly known as the Cambridge Arch as well as over some smaller faults. Also, the low-Ca (high-Na) anomaly of Hocking County lies directly on the Starr Fault System. It is possible that the low-Ca (high-Na) anomaly in northwestern Muskingum County is also correlated with some undiscovered fault or fault system. Finally, the low-Ca (high-Na) in Lake and Geauga counties can be correlated with the Akron Magnetic Boundary.

These fault systems provide the pathways for the Na-rich brines, formed in part by the dissolution of salts of the overlying Salina Formation, thus giving rise to the Na-rich brines of Hocking, Muskingum, and Noble counties.

Dilution of Brines

f_D generally decreases eastward in eastern Ohio. This is expected, considering the geology of eastern Ohio. The sedimentary rocks of Paleozoic age in this portion of the state form a homocline dipping to the east and southeast on the northwest flank of the Appalachian basin (OVERBEY and HENNIGER, 1971). Therefore, the regions in which the Clinton is the shallowest are also the regions in which Clinton brines show the relatively high dilution.

Relationship between f_A and Depth

Figure 8 shows that f_A decreases only slightly as depth increases, indicating that f_A is essentially independent of depth. These findings are quite unlike that reported by LOWRY, 1986. In that study, LOWRY found that the abundance of Na-rich brine decreases with depth and that the Ca-rich brine increases with depth. LOWRY explained this result by the fact that the marine evaporite deposits of the Salina

Formation stratigraphically overlie the Clinton sandstones, while the Ca-rich brine may have a deep-seated origin within the aquifers of the Appalachian mountains (FAURE *et al.*, 1988).

The present results differ significantly from LOWRY'S, due to a large degree of variation in the f_A reported in a number of counties. The counties in question are Summit, Wayne, Holmes, Coshocton, and Licking. LOWRY had nine samples from wells in these counties, with f_A's ranging from 0.54 to 0.67, with an average of 0.62.

In the present study nine samples were from wells in the same counties. The f_A's reported for these samples range from 0.05 to 0.36, averaging 0.17. The relatively low f_A's for these nine samples in the present study have the effect of increasing the verticality of the best-fit line in Figure 8, thus indicating that f_A is essentially independent of depth.

SUMMARY OF CONCLUSIONS

In this study, the chemical variations in brines of the Silurian Clinton sandstone have been shown to be the result of subsurface mixing of two brine types of different chemical composition, a Na-rich brine and a Ca-depleted brine. The brines were also shown to have undergone varying amounts of dilution by groundwater or meteoric water.

The undiluted Na and Ca concentrations were determined for each sample. The interpretation of the data indicates that the concentrations of the Na-rich and Ca-rich brines are virtually constant with depth, although the concentration of the Na-rich brine decreases slightly, while the concentration of the Ca-rich brine increases slightly with depth.

Four high-Na, low-Ca anomalies were identified. These anomalies are

located in northeast Ohio in Ashtabula, Geauga, and Lake counties, and in southeast Ohio in Hocking, Muskingum, and Noble counties. This study has demonstrated that most of these anomalies can be correlated with major basement fault zones, such as the Cambridge Cross-Strike Structural Discontinuity, Starr Fault System, and the Akron Magnetic Boundary.

APPENDIX

Study Number	Sample Number	County/Township	Na (mg/L)	Ca (mg/L)
1	1-85	Mahoning/Berlin	43,100	66,500
2	2,3-85	Muskingum/Springfield	62,900	49,600
3	4,5-85	Holmes/Mechanic	60,900	43,000
4	6-85	Holmes/Richland	33,300	19,600
5	7-85	Perry/Bearfield	60,300	52,300
6	8-85	Portage/Edinburg	64,400	42,400
7	13-85	Wayne/East Union	69,600	38,900
8	15-85	Portage/Freedom	60,900	53,100
9	17-85	Noble/Jackson	78,000	29,300
10	19-85	Coshocton/Adams	61,500	43,800
11	20-85	Stark/Nimishillen	53,800	56,000
12	23-86	Athens/Trimble	71,400	46,300
13	24-87	Licking/Licking	58,300	46,100
14	25-87	Licking/Licking	54,900	45,400
15	26-87	Athens/Trimble	68,300	45,000
16	27-87	Hocking/Falls	61,500	51,000
17	28-87	Licking/Bowling Green	64,300	49,300
18	29-87	Perry/Pike	65,100	48,900
19	30-87	Perry/Harrison	59,800	48,500
20	31-87	Perry/Pleasant	48,900	30,300
21	32-87	Muskingum/Newton	51,800	39,600
22	41-88	Hocking/Green	61,200	47,500
23	42-88	Hocking/Starr	36,600	10,700
24	44-88	Perry/Monday Creek	51,700	32,700
25	45-88	Hocking/Falls Gore	64,600	47,400
26	46-88	Hocking/Washington	38,800	25,700
27	47-88	Perry/Hopewell	63,700	46,800
28	48-88	Muskingum/Muskingum	61,800	48,300
29	51-88	Morgan/Deerfield	73,400	38,400
30	52-88	Muskingum/Jackson	23,800	3,560
31	53-88	Coshocton/Washington	61,000	42,000
32	54-88	Ashtabula/Saybrook	69,900	35,000
33	55-88	Ashtabula/Geneva	71,500	33,600
34	56-88	Ashtabula/New Lyme	67,700	38,900
35	58-88	Trumbull/Bristol	72,600	41,400
36	59-88	Geauga/Parkman	68,200	42,100
37	60-88	Geauga/Montville	80,200	28,700
38	61-88	Geauga/Huntsburg	70,200	40,500
39	62-88	Geauga/Burton	67,600	41,600
40	63-88	Portage/Shalersville	62,800	49,000
41	64-88	Summit/Hudson	64,700	43,000
42	67-88	Lake/Perry	79,900	30,700
43	68-88	Medina/Sharon	52,800	31,900
44	84-88	Muskingum/Rich Hill	34,700	8,190
45	85-88	Muskingum/Meigs	33,700	14,900

Table 1. Original sample numbers, location, and concentrations of Na and Ca in brines from the Clinton Sandstones.

Study Number	Na (mg/L)	Ca (mg/L)	f_A	f_D	Depth (ft)
1	---	---	---	---	5061
2	67,400	53,300	0.09	0.07	3450
3	69,700	49,200	0.18	0.13	3940
4	73,000	43,300	0.32	0.55	3273
5	65,500	56,900	0.01	0.08	3802
6	71,100	46,800	0.24	0.10	4623
7	74,000	41,300	0.36	0.06	3483
8	65,300	57,300	0.00	0.07	4384
9	80,200	30,200	0.62	0.03	5757
10	69,500	49,500	0.18	0.12	4379
11	---	---	---	---	4778
12	71,400	46,300	0.25	0.00	3714
13	67,400	52,900	0.09	0.14	2424
14	66,500	55,000	0.05	0.18	2384
15	71,000	46,900	0.24	0.04	3761
16	66,400	55,200	0.04	0.07	2410
17	68,100	52,100	0.12	0.06	2821
18	68,500	51,400	0.13	0.05	3186
19	66,800	54,400	0.06	0.11	3489
29	72,200	44,700	0.29	0.33	3570
21	68,100	52,100	0.12	0.24	3314
22	67,800	52,700	0.10	0.10	3100
23	83,400	24,500	0.75	0.56	2925
24	71,800	45,400	0.27	0.30	2906
25	69,000	50,500	0.15	0.06	2973
26	70,900	47,000	0.23	0.45	2640
27	68,900	50,700	0,15	0.08	2996
28	67,700	52,700	0.10	0.09	3363
29	74,900	39,700	0.40	0.02	4236
30	89,400	13,500	1.00	0.74	2934
31	70,200	48,300	0.10	0.12	3558
32	75,800	38,200	0.44	0.08	3019
33	76,800	36,200	0.48	0.07	2903
34	73,600	42,100	0.35	0.08	3524
35	73,600	42,000	0.35	0.01	3949
36	72,300	44,600	0.29	0.06	3884
37	80,700	29,000	0.64	0.01	3673
38	73,500	42,400	0.34	0.04	3914
39	72,400	44,500	0.29	0.06	3882
40	67,700	52,900	0.10	0.07	4118
41	70,800	47,200	0.23	0.09	3863
42	79,900	30,700	0.61	0.00	2791
43	72,700	43,800	0.31	0.27	3506
44	85,900	20,000	0.85	0.59	4906
45	78,000	34,300	0.53	0.57	4710

Table 2. Concentrations of Na and Ca with the effects of dilution removed, f_A , and f_D in brines from the Clinton sandstones. These values were not computed for samples 1 and 10 because they may be affetced by a third brine. Also listed are the well depths from which the samples were taken.

		Depth			f_A		
Depth Intervals	Samples	Mean	Standard Deviation	Deviation of the Mean	Mean	Standard Deviation	Deviation of the Mean
2200-2999	10	2725	229.4	72.4	0.219	0.18	0.057
3000-3399	6	3209	120.7	49.3	0.201	0.13	0.053
3400-3799	8	3532	64.9	22.9	0.275	0.18	0.064
3800+	11	4067	259.8	78.3	0.200	0.12	0.036

Table 3. Mean, standard deviation, and deviation of the mean for the depth and f_A for the four depth intervals listed in the table.

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